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Analysis of electroosmotic characters in fractal porous media



Mingchao Liang, Shanshan Yang, Tongjun Miao, Boming Yu*

School of Physics, Huazhong University of Science and Technology, Wuhan 430074, PR China

HIGHLIGHTS

• Fractal model for the height difference caused by the EOFs in porous media is built.

• Fractal model predictions are in good agreement with the experimental data.

• Factors influencing the height difference are discussed and analyzed in detail.

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ABSTRACT

Electroosmotic flow (EOF) in porous media has very important applications in various scientific and engineering fields. In this article, a fractal model for the height difference caused by the EOFs in porous media is developed based on the fractal theory of porous media and on the electrokinetic flow in capillaries, whose tortuosity is taken into account. The proposed model indicates that the height difference is a function of the physical properties of electrolyte solutions, micro-structural parameters of porous media, zeta potential on the solid surface and Debye thickness near the solid–liquid interface in porous media. Factors influencing the height difference are also analyzed. The model predictions show the same variation tendencies with the available experimental data reported in literature. It is found that the proposed fractal model is superior to the hydraulic radius model in terms of dealing with transport in porous media.

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1. Introduction

Electroosmotic flow (EOF) is a ubiquitous electrokinetic effect in many fields such as petroleum engineering, soil physics and chemical engineering. Since Reuss (1809) first discovered the electrokinetic effects in an experiment on porous clay in 1809, many scholars have studied the effects in microchannels and porous media theoretically and experimentally.

An analytical expression for the electrokinetic flow in a narrow cylindrical capillary was first derived by Rice and Whitehead (1965) based on the Poisson and Navier–Stokes equations and the Boltzmann approximation. Moreover, this theory was extended to high surface potentials in fine cylindrical capillaries by some researchers (Levine et al., 1975; Olivares et al., 1980). Later, Ohshima and Kondo (1990) developed a theory for the electrokinetic flow between two parallel similar plates. Mala et al. (1997) also used the microchannel between two parallel plates to investigate the interfacial electrokinetic effects on characteristics of liquid flow by experiment and theory and discussed the effects of the Electric Double Layer (EDL) field, the ionic concentration and the channel size on the velocity distribution and friction factor. Furthermore, the EDL free energy has huge impact on the wettability by triggering a hydrophilicity-inducing tendency, which in turns makes originally hydrophilic surface to a superhydrophilic state, and an originally hydrophobic condition leads to a less hydrophobic state (Das et al., 2012). In addition, Chen et al. (2004) presented a mathematical model by applying the general Nernst-Planck equation, the Poisson equation and the modified Navier-Stokes equation, and they studied the electrokinetic effect on liquid developing flow in a parallel slit. Electroviscous effects were often predicted in steady state, pressure-driven liquid flow in a slit-like (Davidson and Harvie, 2007) or cylindrical microfluidic (Bharti et al., 2008) contraction at low Reynolds number based on a finite volume numerical method. Das et al. (2013) also studied the variations of the streaming potential and electroviscous effect in a nanocapillary with thick overlapping EDLs and observed that the electroviscous effect varies with the streaming potential at small EDL values. However, the electroviscous effect is independent of streaming potential for overlapped EDL. Barz (2009) presented a comprehensive model for electrokinetic flow and transport of electrolytes in microchannels with conductivity gradients. Choi and Kim (2009) investigated the electrokinetic flow

^{*} Corresponding author. Tel.: +86 27 8754 2153. *E-mail address:* yubm_2012@hust.edu.cn (B. Yu).

and ion transport through a silica nanofluidic channel filled with electrolytes experimentally and numerically and found good agreement between model predictions and experimental data. Dutta (2013) applied the advantage of a pressure-gradient in combination with a counteracting electroosmotic flow field to theoretically study the nanofluidic separation of non-neutral analytes.

Brož and Epstein (1976) employed dilute solutions of potassium iodide in purified water as the electrolyte to investigate electrokinetic flow through two types of porous media. Later, Auriault and Lewandowska (1994) analyzed the simultaneously electrical and water fluxes in porous media and presented coupled macroscopic transport equation. Study of the coupled phenomenon, over the last couple of decades, has attracted many researchers' considerable attention. The cylindrically and annularly geometrical models for electroosmotic flow rates through porous media were proposed by Wu and Papadopoulos (2000). Moreover, differences in the predictions from the two models were compared and discussed based on the electrokinetic remediation of contaminated soil. Recently, Pascal et al. (2012) analyzed the effect of capillary geometry on predicting electroosmotic volumetric flow rates in porous or fibrous media by using capillary bundle model. The electrokinetic wall effect caused by different values of the zetapotential associated with the inner surface of the capillary and those of the particles was studied experimentally (Tallarek et al., 2001) and mathematically (Kang et al., 2004), respectively. Apart from this, Bandopadhyay and Chakraborty (2012) reported that the effects of interfacial permittivity and finite ionic sizes affect the electrokinetics significantly in nanochannels. Schaefer and Nirschl (2010) examined the influence of the formation of the electrochemical double layers on the solid-liquid interfaces on electro hydrodynamic transport in nanoporous packed beds. In addition, some researchers (Luong and Sprik, 2013; Paillat et al., 2000) performed experimental investigations on the electroosmosis in porous media and used a theoretical model to explain the electrokinetic phenomenon. The electro-permeability tensor (Bandopadhyay et al., 2013), which characterizes the electroosmotic flow through composite porous media, was obtained by using the Mandelbrot set (Mandelbrot, 1967) and method of homogenization, and they comprehensively analyzed the different representative volume elements, which have fractal boundaries. Hamzehpour et al. (2014) applied a network of spatially distributed rectangular microchannels with random orientations to investigate the electroosmosis phenomena in a two-dimensional (2D) disordered porous medium.

It is well known that the pore structure of many porous media, such as soil, randomly packed beds and fibrous media usually has extraordinary complexity with pore sizes extended over several orders of magnitude. Furthermore, it has been shown that the pore sizes of porous media satisfy statistical fractal characters (Katz and Thompson, 1985; Krohn and Thompson, 1986; Feder and Aharony, 1989; Yu and Cheng, 2002; Yu et al., 2002; Feranie et al., 2011), and the fractal geometry theory for porous media has successfully been applied to analyze transport properties (Cai and Yu, 2011; Cai et al., 2012; Zhu et al., 2012; Cai and Sun, 2013; Zheng et al., 2012, 2013; Xiao et al., 2012, 2014; Liang et al., 2014a, 2014b) in porous media.

Although many scholars have studied the EOF in porous media through experimental research, numerical simulations and theoretical analysis, analytical model is still desirable to be able to further elucidate the effects of the complexity of porous media on the EOF. In this work, analytical model for the height difference caused by the EOF in porous media is derived based on the fractal geometry theory for porous media and the capillary model.

2. The existing model of the height difference caused by EOFs in porous media

In a *U*-tube experiment, Reuss (1809) found that the electrolyte solution rose on one side and lowers on the other side in the

experimental setup (see Fig. 1; Paillat et al., 2000) when the electric voltage was applied across the porous medium saturated with the electrolyte solution. This experiment indicates that there exists the height difference $\Delta h(t)$. Based on the electroosmotic flow in a single capillary (Rice and Whitehead, 1965), the hydraulic radius model for the height difference $\Delta h(t)$ caused by EOFs in porous media is given by (Paillat et al., 2000)

$$\Delta h(t) = \Delta h_{h \max} \left[1 - \exp\left(-\frac{t}{\eta_h}\right) \right]$$
(1a)

where the maximum height difference $\Delta h_{h \text{ max}}$ is expressed as (Paillat et al., 2000)

$$\Delta h_{h \max} = \frac{8\varepsilon |\zeta| V_A}{\rho g r_h^2} \left[1 - \frac{2I_1(\kappa r_h)}{\kappa r_h I_0(\kappa r_h)} \right]$$
(1b)

where κ is the reciprocal of the double-layer thickness (Wang et al., 2006) near the wall in a capillary, I_0 and I_1 are the zero-order and first-order modified Bessel functions, respectively. Generally, the hydraulic radius r_h (its scale is micron) is far larger than the double-layer thickness $1/\kappa$ (Rice and Whitehead, 1965) (its scale is nanometer; Paillat et al., 2000). Thus, $\kappa r_h \gg 1$, and then the term $(2I_1(\kappa r_h)/[\kappa r_h I_0(\kappa r_h)] \sim 10^{-3})$ from Eq. (1b) tends to 0. So Eq. (1b) is simplified as

$$\Delta h_{h \max} = \frac{8\varepsilon |\zeta| V_A}{\rho g r_h^2} \tag{1c}$$

In Eqs. (1a)–(1c), ε and ρ are the dielectric constant and density of electrolyte solution, ζ is the zeta potential (Nägele, 1989) of solid surface, V_A is the tangential electric potential, g is the gravitational acceleration, and η_h is the response time. Furthermore, η_h and r_h are determined by the following relations (Glover and Walker, 2009; Luong and Sprik, 2013):

$$\eta_h = \frac{4\mu R^2 L_0}{A_c N_h \rho g r_h^4} \tag{2}$$

$$r_h = \sqrt{\frac{8K}{\phi^b}} \tag{3}$$

with *K* being the permeability of porous media, ϕ and L_0 being the porosity and length of porous media, μ being the viscosity of electrolyte solution, *R* being the radius of the tubes in both sides of the experimental setup (Paillat et al., 2000) (Fig. 1), A_c being the cross-sectional area of a porous medium, N_h being the average number of pores per cross-sectional area in porous media, and b(=3/2) being empirical constant. However, the hydraulic radius r_h was calculated by



Fig. 1. Schematic of experimental setup (Paillat et al., 2000) for electroosmotic flow through a porous medium.

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